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THE INTERACTION OF ELECTROMAGNETIC RADIATION WITH SOLID MATERIA--ETC(U)
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Final Report for AFOSR Grant No. 76-2887 (9/1/75 - 12/31/77)

Grant title: The Interaction of Electromagnetic Radiation with Solid Materials

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I. Introductory Remarks

The solid state theory effort of the two co-principal investigators has received its primary support from funds provided by AFOSR, under Grant No. 76-2887. The grant period has been an active and productive one for the group. Research completed under previous AFOSR grants has attracted the attention of a number of postdoctoral researchers who have generated their own support to join our group. Thus has produced a theory group of substantial size during the grant period covered in the present report.

We have completed research in most of the problems delineated in our annual proposal, and on a number of other related questions as well. Our earlier progress reports have discussed the individual papers in detail, with the exception of a fraction of them completed during the final six month grant period. In this report, we provide an overview of the completed research, with emphasis on the interrelationships between the various specific topics we have explored. We break down the major research areas into four broadly based subareas for this purpose.

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II. Summary of Completed Research

(a) Surface and Size Effects on the Excitation Spectra of Solids, and on the Interaction of Electromagnetic Radiation with Crystals from Microwave through Visible Frequencies

For many years, the study and analysis of the interaction of electromagnetic radiation with solids has proved a most powerful method of probing a wide variety of physical processes in solids. In recent years, one has come to appreciate that a wide variety of electromagnetic phenomena are associated with surfaces and interfaces. In essence, many of the fundamental interaction processes studied in the bulk may also be studied on the surface, through use of waves (surface polaritons) with electromagnetic fields localized near the surface. Just as in the corresponding bulk studies, one may use these waves to learn about the physics of crystal surfaces or interfaces between different materials. Also the fundamental physical properties of these waves are quite fascinating and frequently unusual, and they prove potentiallly useful for applications.

We have had a strong interest in the study of the basic properties of the surface polaritons, the nonlinear interactions in which they participate and the methods for probing them. This interest leads one to inquire about the basic physics of the crystal surface region, with emphasis on its influence on the interaction of electromagnetic radiation with solids. More recently, in collaboration with Professor Ushioda, a joint theoretical and experimental study of size effects on

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nonlinear interactions (light scattering) between laser radiation and polaritons (both surface and bulk) has been completed. We turn here to a review of the research completed in these areas.

During the grant period, we completed two series of studies of nonlinear interactions between electromagnetic waves near and on surfaces and interfaces.

One of these[(1),(2)] is a study of the scattering of light from surface and interface polaritons, i.e. electromagnetic waves that propagate along the surface or an interface, with electromagnetic fields localized near the surface. This effort produced a quantitative theory[(1)] of the line intensities measured by Ushioda and co-workers, along with an explanation of the failure to observe surface waves in the conventional back scattering geometry. A second and more complete description of this phenomenon led to rather straightforward but technically complicated formulas for the total spectrum of light scattered from a film on a substrate, with the surface (and interface) polariton features as well as size dependent effects in the bulk polariton fully included.[(2)] This work was coordinated with experimental studies in Professor Ushioda's laboratory, and provides a quantitative theory of their results.

We have had a major success in this area very recently. Motivated by the fabrication of very thin, free standing films in Ushioda's laboratory, we applied the light scattering theory to the analysis of Raman scattering of films in the 5 μm to 50 μm thickness range.[(3)] The theory produces the whole Raman spectrum, and not just line intensities. We were startled

to see the theory predict very strong scattering from well resolved "guided wave polariton modes" in these films. The theory outlined rather precisely the conditions that allows the modes to be seen, and they were found in the laboratory at the same time.[4] These modes are the analogue, for infrared frequencies, of the guided waves that form the basis of integrated optics technology.[5] Our joint theoretical and experimental proposal constitutes the first study of these waves in the infrared, and in a spectral region with strong dispersion in the dielectric constant of the film.

The above study examines the nonlinear interaction between an external electromagnetic wave (the laser photon) and either surface polaritons, or bulk polaritons in a confined geometry. If a surface polariton is launched, it can also mix with a second such wave to produce output radiation at the sum or difference frequency either in the form of a surface polariton, or a bulk wave depending on the kinematics of the mixing process. One can envision studies in "nonlinear surface optics" analogous to nonlinear optics in the bulk. Studies of such nonlinear interactions between surface polaritons were initiated by Maddox and Mills,[6] and a subsequent more detailed analysis has been completed by Bonsall and Maradudin.[7] The calculations by Bonsall and Maradudin outline in some detail quantitative features of the nonlinear coupling, at infrared frequencies for a wide range of kinematical parameters. A by-product of this investigation is the development of a quantitative theory of the nonlinear subsceptibilities

of GaAs, at frequencies appropriate to microwave or infrared studies.

The interest in electromagnetic response of the surface region has led us to explore a number of other phenomena in this area. For example, the image potential experienced by a moving charge near a surface has its origin in the coupling between the charge and surface polaritons, in one simple microscopic description of this quantity. One of us[8] has constructed a rather general description of the image force experienced by a moving charge particle, to correct an erroneous assertion in the literature that at large distances from the surface, retardation effects play a crucial role.

The above descriptions of the optical response of the surface assume that the surface region is described by the same local dielectric tensor as the bulk of the crystal. In many circumstances, it is not obvious this presumption is reasonable. Indeed, a fascinating possibility is the exploitation of the waves and probing methods outlined above to explore deviations of the dielectric response from its bulk character near the surface. A series of studies have been carried out which explore the influence of such deviations on the electromagnetic response of the surface.

It is known that in some regions of frequencies, most importantly just below the absorption edge of semiconductors near exciton absorptions, it is a poor approximation to suppose the bulk dielectric constant is local in character,

that is that the dipole moment per unit volume $\vec{P}(\vec{x})$ is proportional to the electric field $\vec{E}(\vec{x})$ at the same point in space. In these frequency regions, nonlocal effects ("spatial dispersion" effects) become very important. In bulk measurements, in spectral regimes where spatial dispersion is important, the surface region exerts an important effect on observation through its influence on the form of the "additional boundary condition" that enters the theory. We have carried out a series of studies which outline the possible form this boundary condition may assume when certain constraints are imposed,[9] and the manner in which the precise form of the additional boundary condition influences a variety of optical measurements.[10]-[12]

Nonlocal effects are important not only at visible frequencies, but also dominate the surface response of metals at microwave frequencies. We have carried out a detailed study of the contribution from electron-phonon scattering to the surface impedance Z of metals in the highly non-local (anomalous skin effect) regime.[13] For a simple model, we outline the temperature variation of Z in some detail, to lowest order in the electron-phonon scattering rate. The question addressed here is whether the relaxation time which enters the surface impedance is identical to that which enters the D.C. electrical resistivity. We find the answer to be negative, although the temperature variation of the two scattering rates are similar.

We conclude this subsection with mention of two final topics we have explored in the above area. This is the propagation of electromagnetic waves on a semiconductor surface with an inversion layer present. This is an example of a situation where the wave may be used to explore the physical properties of this fascinating two-dimensional inversion layer; in a material such as silicon, the surface polariton is "bound" to the surface only by virtue of the free carriers present in the inversion layer. Furthermore, the electric field in the surface polariton assumes its largest value at the interface where the inversion layer is present; the propagation characteristics of the wave are thus sensitive probes of the inversion layer.(14) Finally, surface roughness affects both the frequency and attenuation length (lifetime) of surface polaritons. A theory which gives the roughness-induced frequency shift and attenuation length has been developed.(15)

(b) <u>Surface Wave Propagation at Microwave Frequencies</u>; <u>Rayleigh Waves</u> (Surface Acoustic Waves)

At infrared frequencies, the surface polaritons which propagate on surfaces have penetration depths the order of a micron, in typical cases. At microwave frequencies, although the frequency is very much lower than the infrared, the small value assumed by the sound velocities of solids (compared to the velocity of light) insures that the penetration depth and wave length of Rayleigh waves is also the order

of a micron. Thus, Rayleigh waves serve as a probe of the physics of the surface region every bit as well as surface polaritons. Because of their much lower frequency, however, one can envision exploring very different physics with Rayleigh waves.

During this past grant period, we have explored the sensitivity of Rayleigh waves to perturbations of the surface region. For example, we have developed a theoretical description of the attenuation of these waves by roughness on the surface.[16] The mathematical apparatus constructed in the course of this study will prove useful in a variety of theoretical studies of Rayleigh waves and their interactions.

Another area we have pursued is the propagation of Rayleigh surface waves which propagate on a ferromagnetic substrate. One finds strong attenuation of the Rayleigh wave when its frequency lies either in the frequency band where bulk spin waves exist, or when its frequency matches that of the Damon-Eshbach surface spin wave. We have been intrigued by this phenomenon, because the Rayleigh waves may prove an extremely useful microwave frequency probe of the response of magnetic materials near their surface. We believe these waves may be used on surfaces characterized more precisely than those employed in ferromagnetic resonance experiments of thin films. At the same time, since the spin wave frequencies of the substrate can be tuned through variation of the magnitude of an external magnetic field, the propagation characteristics of the Rayleigh wave may be manipulated

at will, with strong field dependent attenuation or dispersion introduced in this way.

The interest in microwave studies of the response of ferromagnets generated through the Rayleigh wave studies has led us to think more carefully about the nature of magnetic order at surfaces. We find an interesting magnetic instability (dubbed "magnetic surface reconstruction") at the surface[18],[19] which can produce pinning fields there that affect the magnetic field[20] and temperature variation[21] in the microwave response of a magnetic film. Rather strong evidence that the instability we predict actually occurs comes from experimental studies of ferromagnetic resonance in EuS films (thickness of a few hundred Angstroms) deposited on a quartz substrate. The measurements show an anomalous temperature variation of an effective pinning field experienced by the surface spins; such an anomalous temperature variation is expected in the presence of magnetic surface reconstruction.[21]

(c) Theory of Wave Propagation on Nonplanar Surfaces; Edge Modes, Rounded Corners and Topographic Waveguides

The analyses of wave propagation and nonlinear interactions on or near surfaces described in the preceding two subsections all concern perfectly flat plane surfaces or interfaces, with possibly small amplitude roughness present. A whole new class of modes, acoustical and electromagnetic, can arise near edges or features on crystal surfaces. By modern fabrication techniques one may cut grooves in surfaces,

or create ridges which run along the surface. Such linear geometrical structures serve to guide waves localized in their near vicinity ("topographic waveguides"). We have been engaged in a series of studies of wave propagation along linear features such as those described above. Before we describe the specific calculations carried out in this phase of our activity, we begin with some general comments.

The general structure of the mathematical theory of surface wave propagation (acoustical or electromagnetic) along a planar surface is by now well known, although in any particular case it may prove difficult to carry through a concrete calculation to the end. If one has a field amplitude $u_i(\vec{x},t)$, with $\vec{x}_\parallel$ the projection of $\vec{x}$ onto a plane parallel to the surface and $\hat{z}$ the direction normal to it (with the crystal in the half space $z > 0$), then one writes

$$u_i(\vec{x}t) = \sum_j u_i^{(j)} \exp[i\vec{k}_\parallel \cdot \vec{x}_\parallel - \alpha_j z - i\Omega t] \qquad (1)$$

where bulk equations of motion (Maxwell's equations, equations of elasticity) determine the attenuation constants α_j, and the ratios $u_i^{(j)}/u_\ell^{(k)}$. One submits the above solution to the boundary conditions to obtain a relation between Ω and $\vec{k}_\parallel$ -- the dispersion relation of the surface wave.

Near a linear feature on the surface, one has translational invariance in only one spatial direction. If we call this the x direction, Eq. (1) is replaced by

$$u_i(\vec{x},t) = u_i(y,z) \exp(ik_\parallel x - i\Omega t) \qquad (2)$$

where only in special cases can $u_i(y,z)$ be expressed in terms of elementary functions.

Before one approach the study of wave propagation in a particular case, it is necessary to develop a tractable mathematical method to treat complex forms such as that in Eq. (2). A major accomplishment of our program has been the successful development of procedures to study these waves within the framework of a computationally tractable scheme that allows convergence of the necessarily approximate calculations to be checked at each stage. With these procedures, we have carried out a series of accurate studies for the realistic geometries outlined below. We wish here to emphasize the importance of erecting a useful and practical scheme for carrying out calculations of this class; these schemes may well prove of wide utility in the study of excitations in finite crystals.

During the current grant period, our earlier studies have been extended to a more complete description of the edge localized vibration modes that propagate on a ridge of rectangular cross section.[22] At low frequencies, the waves are found to involve coherent motion of the ridge as a whole, while when the wavelength is small compared to the width of the ridge, some of the modes evolve into edge modes which propagate along the two edges.

We have also carried out the first studies of propagation near edges and for rectangular bars, for a gyrotropic medium (ferromagnets). These calculations show a rich and

varied mode structure.[(23),(24)]

The calculations carried out to date all concern modes in the vicinity of an idealized structure with perfectly sharp corners. During the present grant period, the effect of a finite radius curvature of corners was studied through analysis of the electrostatic normal modes of a structure of parabolic cross section.[(25)] The rounded tip of the parabola mimics the finite curvature of real edge; it proved most informative to compare the results of this recent study with earlier ones for the electrostatic modes of a triangular wedge with perfectly sharp apex.

(d) Optical Interactions in Impure and Disordered Solids

A strong and continuing interest of our group is the study of optical interactions (absorption, light scattering) in solids. While the investigations described in subsection (a) explored such processes in the near vicinity of a surface or interface, during the present grant period we actively pursued the theory of optical interactions in the bulk, where surface or size effects are not important. The studies completed in this grant period center primary attention on the influence of defects on optical properties.

We have completed two studies of infrared absorption in the Drude tail of doped (n type) semiconductors.[(25),(26)] A variety of processes which contribute to this free carrier absorption were studied by theorists some years ago. Yet the theory fails to give an adequate account of the data, and a number of important questions remain to be posed.

For example, free carrier absorption in ionic materials can be dominated by a process where an electron absorbs the photon, then subsequently emites an LO phonon via the Fröhlich interaction. (This process does not require an impurity or defect to be present.) The contribution of this process to the absorption rate was calculated a number of years ago. Many of the experiments are carried out at carrier concentrations sufficiently high that the electron plasma frequency lies above that of the LO phonon, yet there is no calculation which takes due account of the coupling between the LO phonon and the electron plasma. Our first study analyzes this regime,[25] to conclude surprisingly that for quantitative purposes the electron plasma has little influence on the calculated absorption coefficient.

A second study[26] examines the free carrier absorption induced by the presence of ionized impurities. Here a photon is absorbed, and the electron recoils by scattering from an ionized impurity. The potential seen by the electron is that from the charge Ze associated with the ionized impurity, and its associated screening charge. We note that the screening charge does not remain static during the scattering event, but may respond dynamically to the incoming electron. In essence, the electron may scatter off the impurity, with the emission of plasmons. We have calculated the absorption constant for such a model, to find clear, pronounced structure at the electron plasma frequency. Such structure has been clearly and unambiguously observed recently by the Balkanski group in Paris.

Another study[27] examines a distinctly different aspect of the interaction of electron plasma oscillations with impurities. In an impure semiconductor, light impurities give rise to localized phonon modes, with frequency higher than the maximum vibration frequency of the crystal. If the impurity has a non-zero dynamic effective charge, then the local mode is infrared active; a macroscopic electric field is set up when the local mode is excited. Such a local mode can couple to the electron plasma motion in a doped semiconductor. This coupling was studied some years ago by Maradudin and Sham, who found it to be extremely weak, for an isolated impurity placed in a doped semiconducting crystal.

Experiments on the $GaAs_{1-x}P_x$ system show that for small x, the P-induced local mode couples strongly to the electron motion. We argue[27] this occurs because for finite concentrations, the impurity array may be excited coherently, to produce a macroscopic electric field that couples strongly to the electron plasma. We have constructed a theory of this coupling, within the framework of a method that reproduces the Maradudin-Sham result in the limit of very low concentration. The effects are illustrated by a calculation of the Raman spectrum of the system. Light scattering experiments on the $GaAs_{1-x}P_x$ system are being set up currently, and we hope to have spectra to compare with our theory in the near future.

Another topic pursued this grant period is the interaction of electromagnetic radiation with Jahn-Teller impurities

in insulators. This work was stimulated by the presence of Dr. E. Mulazzi as a visitor to the group. In the presence of a strong Jahn-Teller effect in a degenerate excited state, optical selection rules are violated, i.e. the selection rules for electronic Raman scattering from the impurity differ distinctly from those appropriate to the rigid lattice. A quantitative description of these selection rule violations has been worked out by our group during the present grant period.[28]

A final topic in this area is the study of the influence of an impurity on the electronic contribution to the absorption coefficient of metals.[29],[30] The emphasis here is on the manner in which an impurity placed in the lattice may "activate" a non-vertical optical transition, leading to new structure in the absorption coefficient absent in the pure material. While numerous experimental studies of optical properties of alloys have been reported in the literature, rather little emphasis has been placed on this possibility. We believe, as discussed in our second publication, that the reason for this is the incorrect quantity has been extracted from the data. Our model calculations show that $\epsilon_2(\omega)$, the imaginary part of the dielectric constant, shows very mild structure hard to perceive even in the theoretical calculations. On the other hand, $\partial\epsilon_2(\omega)/\partial\omega$ shows clear structure at certain characteristic transition energies, with shape dependent on the details of the impurity potential. Thus, it is $\partial\epsilon_2(\omega)/\partial\omega$ that should be studied rather than $\epsilon_2(\omega)$; modulation spectroscopy is clearly the method to be used here.

(e) Comments on Other Papers

In the preceding subsections, we have commented on theoretical analyses carried out during the grant period which fall roughly into four broad areas. These catagories do not exaust the topics explored during the grant period. For example, we have explored the surface specific heat of finite crystals at low temperatures, for crystals of both cubic and hexagonal symmetry.[(31),(32)] These are the first analytic results for the surface specific heat, for other than the isotropic elastic continuum. Also, the static and dynamic properties of two dimensional electron crystals have been explored.[(33)] This work was carried out with electrons adsorbed on the surface of liquid helium (or any other substrate capable of binding electrons via the attractive image potential) in mind. On such substrates, one can envision absorbing electrons at a surface density sufficiently high that a two dimensional Wigner lattice will form.

(f) Concluding Remarks

We have not attempted to outline the detailed content of all the papers completed by our group during the past grant period. Indeed, not all papers published during the grant period (and listed at the end of the present report) have been cited. We have attempted here to present a picture of the overall thrust of the group, and to organize the research activity into broad subcategories. We refer to our earlier progress reports for a more complete description of the contents of many of the papers cited above.

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III. Personnel on Grant 76-2887

Faculty, Postdoctoral Researcher and Research Assistants who received support are:

Prof. A. A. Maradudin	
Prof. D. L. Mills	
Dr. K. R. Subbaswamy	(postdoctoral researcher)
Dr. T. Rahman	(postdoctoral researcher)
R. Sirko	(research assistant)
R. Q. Scott	(research assistant)
T. M. Sharon	(research assistant)
G. Pinas	(research assistant)
M. F. Bishop	(research assistant)
R. Gutierrez-Lee	(research assistant)

The following postdoctoral researchers joined our group, with salary from the source indicated. They contributed directly to the AFOSR program, and in some cases received support from the grant in support of their activities (computing time, supplies and expenses, etc.).

Name	Date of Visit	Source of Salary
C. Demangeat Université Louis Pasteur Strasbourg, France	9/1/75-8/31/76	N.S.F./C.N.R.S. Exchange Program
J. C. Parlebas Université Louis Pasteur Strasbourg, France	11/1/76-5/31/78	N.S.F/C.N.R.S. Exchange Program
Kh. Pashaev Azerbajdzhan State University Baku, U.S.S.R.	10/27/75-4/15/76	I.R.E.X.

IV. Theses Awarded During Grant Period

The following research assistants received their Ph.D. during the grant period:

Name	Thesis Title
R. Q. Scott (June 1976)	"The Propagation of Surface Magneto-elastic Waves on Ferromagnetic Crystal Substrates"
T. M. Sharon (May 1976)	"Topographic Acoustic and Magnetic Crystal Waveguides"
M. F. Bishop (June 1976)	"Studies of the Effects of Spatial Dispersion and Surfaces on Optical and Dynamical Properties of Crystals"
R. Maddox (June 1976)	"Some Properties of Polaritons in Solids"

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Theoretical research has been completed in four areas of study and analysis of the interaction of electromagnetic radiation with solids: ~~a)~~ Surface and Size Effects on the Excitation Spectra of Solids, and on the Interaction of Electromagnetic Radiation with Crystals from Microwave through Visible Frequencies; ~~b)~~ Surface Wave Propagation at Microwave Frequencies; Rayleigh Waves (Surface Acoustic Waves); Theory of Wave Propagation on → next page

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